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㉚ Optical devices.

㉛ An optical device is made of a polymer material comprising (i) polymer portions constituted mainly of aromatic vinyl monomer units and (ii) polyphenylene ether portions; the average molecular weight of the polyphenylene ether portions being from 0.3 to 0.7, expressed in terms of the intrinsic viscosity measured in chloroform at 25°C.

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OPTICAL DEVICES

The present invention relates to optical devices having low birefringence and which are useful, for example, as optical disk substrates for optically recording and reproducing information and as lenses, prisms, etc.

Recently there has been developed the system of recording and reproducing high-density information, e.g. images and/or sounds, by using laser beams, wherein the information is recorded in the form of fine projections or depressions on disk substrates or in the form of changes in optical properties of recording films formed on disk substrates.

Disk substrates for use in the above recording and reproducing systems should be transparent and in addition should have good dimensional stability, high optical homogeneity, and small birefringence.

A plurality of duplicated substrates can be economically produced by using a resin material to form the disk substrate. However, it is widely recognized

that the resin flows during moulding of the disk substrate and the molecules are oriented during cooling so that birefringence is induced. This is a fatal defect for an optical disk substrate.

5 The molecular orientation caused during moulding is inevitable, particularly in injection moulding. From this point of view, a polymer comprising methyl methacrylate is the only resin material of low optical anisotropy for moulding disk substrates.

10 Known polymers comprising methyl methacrylate, however, have the drawback that, because of their high hygroscopicity, substrates formed thereof have inferior dimensional stability, that is the substrate warps and/or twists in highly humid atmospheres. This
15 drawback is described in detail, for example, in Nikkei Electronics (p. 133, issued June 7, 1982). As a result, aromatic polycarbonate resins of low hygroscopicity are in use as materials for acoustic compact disks.

However, it is difficult to decrease birefringence
20 of the moulded substrates formed of aromatic carbonate resins since the resins contain aromatic rings, which have a large anisotropy, in their backbone chains. Decreasing the molecular weight and the conditions of the moulding are under investigation as means by which

birefringence may be reduced. However, since birefringence of the substrate originates from the material itself, it is impossible to consistently produce substrates with small birefringence and it is 5 extremely difficult at present to produce small-birefringent substrates, larger in diameter than acoustic compact disks, by injection moulding.

To improve dimensional stability, which is a drawback of a polymer comprising methyl methacrylate, 10 copolymers comprising methyl methacrylate and aromatic vinyl monomers have been proposed [see, for example, Japanese Laid-Open Patent Applications Nos. 33446/82, 162135/82, and 88843/83]. However, these copolymers containing aromatic rings are not of practical use because of their liability to large birefringence. For 15 the disk substrates which are used not only for reproducing but also for the recording of information, a material is required which has small birefringence and good dimensional stability, but no resin has yet been found out that can adequately satisfy these requirements.

20 With respect to other optical devices including lenses and prisms, for which methacrylic resins and the like have been used as materials, there are needs for optical devices formed from a resin which is of reduced birefringence and superior in heat resistance, 25 mechanical strength, and dimensional stability.

U.S. Patent No. 4,373,065 discloses an optical recording device comprising an optically isotropic composition prepared by mixing two resins which have opposite optical anisotropies but are completely miscible with each other, in such a ratio that their optical anisotropies will compensate each other so as to reduce the overall birefringence to substantially zero. This patent further describes mixtures prepared by using a polyphenylene ether and a polystyrene as component polymers having opposite optical anisotropies and shows that a film prepared from a mixture such that the anisotropies compensate each other does not cause birefringence even when stress is exerted on the film in the solid state. However, this patent does not reveal that an optical device, such as an optical disk substrate, when formed from the above polymer composition by injection moulding, exhibits small birefringence.

We have found that the birefringence of mouldings, such as optical disk substrates, is not always small when these mouldings are formed by injection moulding (which is recently the most common method of forming optical devices), even from a mixture of two polymers which have opposite optical anisotropies in the solid state and are completely miscible with each other and

wherein the mixing ratio is such that the optical anisotropies in the solid state just compensate each other. That is, it has been found that optical materials, such as an optical disk substrate having 5 small birefringence, cannot be obtained by injection moulding of any composition or polymer mixture chosen simply by considering the optical anisotropy of each polymer in the solid state.

In recent years, attempts have been made to produce 10 plastic substrates for magneto-optical disks where the erasing and rewriting of information are possible.

Information recorded on such an optical disk of the magneto-optical type is read by focussing a polarized 15 laser beam through a lens onto the recording medium of the disk and detecting the Kerr effect-induced slight rotation of the polarization plane of the laser beam reflected from the recording medium. Accordingly, it is necessary in this case to use an optical disk substrate 20 which scarcely causes the birefringence for obliquely incident beams as well as for normally incident beams. In addition, the optical disk substrate should have high heat resistance since the substrate during information writing is heated by the laser beam.

It is an object of the invention to provide an optical device which has small birefringence for vertical incident beams as well as obliquely incident beams even when formed by injection moulding.

- 5 compression compound or the like, and has high heat resistance, well-balanced mechanical strengths, and good dimensional stability.

Basically, the optical device of the invention is formed of a polymer material comprising (i) polymer 10 portions derived mainly from aromatic vinyl monomers, and (ii) polyphenylene ether polymer portions. The optical devices of the invention may take the form, for example, of magneto-optical disk substrates, lenses and prisms, besides common optical disk substrates.

- 15 The polymeric material of the optical device may be a mixture of a polymer derived mainly from aromatic vinyl monomers and a polyphenylene ether; a block or graft copolymer comprising both vinyl aromatic monomer-based blocks and polyphenylene ether blocks; or 20 a mixture of them.

The polymer derived from aromatic vinyl monomers (a polymer constituted mainly of aromatic vinyl monomer units) may be a vinyl aromatic homopolymer or a copolymer comprising at least 50% by weight of aromatic

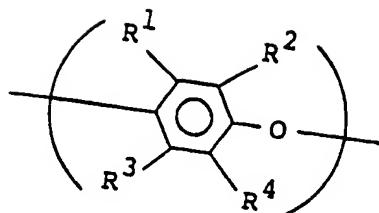
vinyl monomer units. Examples of aromatic vinyl monomers include styrene, α -methylstyrene, m-methylstyrene, p-methylstyrene, o-chlorostyrene, m-chlorostyrene, p-chlorostyrene, m-bromostyrene and p-bromo-
5 styrene. In particular, styrene is favourably used.

Suitable monomers for copolymerization with such aromatic vinyl monomers include; unsaturated nitriles, e.g. acrylonitrile and methacrylonitrile; alkyl methacrylates, e.g. methyl methacrylate, n-propyl
10 methacrylate, isopropyl methacrylate, n-butyl methacrylate and cyclohexyl methacrylate; alkyl acrylates, e.g. methyl acrylate, ethyl acrylate, propyl acrylate, and butyl acrylate; and, further, methacrylic acid, acrylic acid, maleic anhydride, citraconic
15 anhydride, N-methylmaleimide, and N-phenylmaleimide. These comonomers may be used alone or in admixture, the type and proportion of the monomers used being suitably chosen within the range thereof where none of them impair the transparency of the resulting polymer
20 composition comprises the aromatic vinyl copolymer and the polyphenylene ether. The amount of aromatic vinyl monomer in the monomer mixture to form the copolymer should be at least 50% by weight, otherwise the resulting resin will have an undesirably high
25 hygroscopicity.

The melt fluidity of the aromatic vinyl polymer expressed in forms of melt flow rate (MFR) at 230°C under 3.8 kg load, is desirably from 0.5 to 200, preferably from 2 to 100. If the MFR exceeds 200, the 5 polymer is poor in mechanical strength and when the MFR is less than 0.5, the birefringence is difficult to reduce.

The aromatic vinyl polymer may be produced by a bulk, suspension, emulsion, or solution polymerization 10 method using a radical initiator, but bulk polymerization and suspension polymerization are preferred in view of the productivity and reduced contamination of the polymer. Suitable radical initiators include peroxides, e.g. lauroyl peroxide, 15 benzoyl peroxide, di-tert-butyl peroxide, and dicumyl peroxide; and azo compounds, e.g. 2,2'-azobisisobutyronitrile and 1,1'-azobis(1-cyclohexanecarbonitrile). If necessary, to control the molecular weight, a chain transfer agent may be added, such as tert-butyl, 20 n-butyl, n-octyl, n-dodecyl, or tert-dodecyl mercaptan. The polymerization is generally carried at a temperature of from 50 to 150°C.

The polyphenylene ether may be represented as a polymer consisting of recurring units of the formula:



in which R¹, R², R³ and R⁴ are the same or are different and each is a hydrogen or halogen atom or a hydrocarbyl group.

5 Polyphenylene ethers, as defined above, are readily prepared by known methods (see, e.g. Japanese Patent Publications Nos. 18692/61 and 36518/72), that is by oxidative coupling of phenolic monomers in the presence of a copper or manganese catalyst.

10 Examples of suitable polyphenylene ethers include poly(2,6-dimethyl-1,4-phenylene) ether, poly(2-methyl-6-ethyl-1,4-phenylene) ether, poly(2-methyl-6-propyl-1,4-phenylene)ether, poly(2,6-dipropyl-1,4-phenylene)ether, and poly(2-methyl-6-bromo-1,4-phenylene)ether. In particular, poly(2,6-dimethyl-1,4-phenylene)ether is preferred.
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Polyphenylene ether resins generally in use as engineering plastics may be acceptable but resins of lower molecular weights are preferred. That is, when the average molecular weight of the polyphenylene ether 5 is expressed in terms of the intrinsic viscosity of the polymer in chloroform at 25°C, acceptable values are 0.1 to 1.0, but desirably the value is from 0.3 to 0.7, preferably from 0.3 to 0.45, and particularly preferably from 0.35 to 0.42. If the value is less than 0.3, the 10 resulting optical device will have poor mechanical strength.

To decrease the birefringence, particularly for obliquely incident beams, the intrinsic viscosity of the polyphenylene ether is desirably up to 0.45.

15 This is conceivably because the vinyl aromatic polymer and the polyphenylene ether, during moulding (e.g. injection moulding) to form an optical device have different tendencies to molecular orientation and/or different relaxation speeds in a mould.

20 Thus, polyphenylene ethers, in moulding, exhibit inherently high melt viscosity and tend to develop molecular orientation, which is liable to remain after moulding; but when a polyphenylene ether of lower average molecular weight is used for moulding an optical

device, the orientation of the polyphenylene ether molecules in the resin mixture will occur with more difficulty and the relaxation speed will higher; therefore optical devices exhibiting reduced 5 birefringence to vertically incident as to well as oblique beams can be obtained from a wider range of resin compositions under wider ranges of injection moulding conditions.

The resin material for the optical device may be 10 prepared by mixing the vinyl aromatic polymer and the polyphenylene ether by a melt or solution mixing technique.

Melt mixing may be carried out using a mixing machine such as an extruder, Banbury mixer, kneader 15 blender, or heated roller mill under high shear stress at temperatures of at least the glass transition point of the polyphenylene ether. The degree of mixing should desirably be such that particle sizes about 1 μ or less, preferably of molecular order. When 20 molecular-order dispersion is reached can be readily judged by observing that the mixture exhibits a single glass transition temperature.

To achieve satisfactory mixing, the mixing temperature may be raised, the mixing period may be 25 extended and/or the shear stress may be increased.

In melt mixing, a small amount of an organic solvent may be used as a plasticizer to reduce the glass transition temperature of both polymers, thereby facilitating mixing. Organic solvents, as noted below, suitable for 5 solution mixing can be used in this case. The solvent may be removed by distillation after completion of the mixing.

Solution mixing may be effected by dissolving each polymer in an organic solvent to a concentration of at 10 least 1% by weight, mixing the solutions with stirring to form a uniform mixture, and either distilling the mixture to remove the solvent or adding a poor solvent for the polymers to the mixture to precipitate the polymers.

15 Suitable solvents for solution mixing include, for example, chloroform, methylene chloride, ethylene chloride, toluene, benzene, and chlorobenzene. Suitable poor solvents include, for example, methanol, ethanol, propanol, n-hexane, and n-pentane.

20 A block of graft copolymer comprising vinyl aromatic polymer and polyphenylene ether blocks can be prepared by polymerizing the monomer(s) to form one of the polymers in the presence of the other polymer. For instance, a block or graft copolymer can be produced 25 according to the methods described in Japanese Patent

Publications Nos. 22069/67, 1210/72, 47862/72, and
38596/77, by either polymerization of a monomer mixture
composed mainly of an aromatic vinyl monomer in the
presence of a polyphenylene ether, or by oxidative
5 coupling polymerization of an appropriate phenolic
monomer in the presence of the vinyl aromatic polymer.

The polymer material forming the optical device of
the invention suitably comprises from 30 to 70% by
weight, preferably from 40 to 55% by weight, of the
10 vinyl aromatic polymer and from 70 to 30% by weight,
preferably from 60 to 45% by weight, of the
polyphenylene ether. If the amount of polyphenylene
ether is less than 30% by weight or more than 70% by
weight, the birefringence of the optical device is not
15 sufficiently small. Further, if the amount of
polyphenylene ether is less than 30% by weight, the
optical device has insufficient heat resistance.

The resin composition of the optical device, in
practice, is suitably chosen within the above defined
20 ranges depending on the method of moulding. In
injection moulding, for example, the resin proportions
may be suitably chosen depending on preset conditions of
moulding, such as resin temperature, injection pressure,
and moulding temperature so that the birefringence of
25 the mouldings will agree with the level required for the
intended purpose of the product.

Optical devices, particularly optical disk substrates, of the present invention in certain applications transmit semiconductor laser beams or the like. Accordingly, the light transmittance of optical 5 devices for such applications is desirably at least 75% at a wavelength of 800 nm when the thickness of optical device is 1.2 mm.

Of the optical devices of the present invention, optical disk substrates can be formed by injection 10 moulding, compression moulding and injection-compression moulding. The effect of the present invention is the more remarkable when a moulding method, among the above, is used which tends to form higher birefringent products, though injection moulding is best suited in 15 view of productivity.

Basically injection moulding comprises heating a resin to a fluid state, forcing the resin into a closed mould cavity, and solidifying the resin by cooling. This simple injection moulding method may be combined 20 with a mould cavity evacuating method or an injection-compression moulding method may be used.

When an optical device of the invention is produced by injection moulding, a melted and plasticized resin is

suitably injected at a temperature of from 270 to 350°C, preferably from 300 to 340°C. This temperature is that of the resin, in the injection cylinder, which has been melted and plasticized by heating with external heaters
5 and with shear heat generated by screw revolution in the injection moulding machine. If the resin temperature is below 270°C, the resulting optical disk substrate may be unsuitable for its intended use since the degree of birefringence may become 20 nm or more. If the resin
10 temperature exceeds 350°C, the resin decomposes, develops defects such as burnings and silver flow marks and bit errors on the resulting optical disk substrate will markedly increase.

In injection moulding, the mould temperature is
15 suitably maintained at a temperature of from 50 to 140°C, preferably from 80 to 120°C. (The mould temperature is the surface temperature of the mould cavity immediately before injection). If the mould temperature is below 50°C, the reproduction of finely
20 engraved grooves on the mould cavity surface deteriorates and if the mould temperature exceeds 140°C, releasability of the moulded resin from the mould becomes worse. The injection period is suitably from 0.2 to 3.0 seconds, preferably from 0.3 to 2 seconds.
25 (The injection period is the perio of time taken to filling the resin into the mould cavity). Injection

periods below 0.2 second may give rise to silver flow marks and may result in increased bit errors when the mouldings are used as an optical disk. Injection periods of longer than 3 seconds undesirably may cause 5 the resulting optical disk substrate to exhibit high birefringence, corresponding to 20 nm or more.

In order that the invention may be well understood the following Examples are given by way of illustration only. In the examples, all parts and percentages are by 10 weight unless otherwise stated.

The physical properties noted in the examples were measured as follows:

Birefringence: Measured using a polarization microscope, the retardation was measured at a wave-length of 546 nm according to the Sernarmont compensator method.

Water absorption: Equilibrium water absorption in water at 60°C was measured in accordance with ASTM D-570.

Light transmittance: The transmittance of a 1.2-mm thick specimen was measured at a wavelength of 800 nm using a self recording spectrophotometer Model 25 330 supplied

According to the invention, it is possible to provide an optical device which comprises a resin of the polystyrene family as an ingredient, which resin being of reduced hygroscopicity and good in dimensional stability but impracticable up to now because of its high birefringent properties. In addition such a resin has a good balance of heat resistance and mechanical strength.

The optical device also comprises a specific polyphenylene ether having relatively low intrinsic viscosity as another ingredient, thereby having low birefringence for vertically incident beams as well as unprecedently for obliquely incident beams, and additionally has wide composition latitude, hence offering high heat resistance.

The optical device of the present invention also has the advantage making it possible to mould low birefringent mouldings even by injection moulding which is generally regarded as producing large birefringence.

Optical devices of the invention include optical disk substrates, lenses, prisms, etc. and are particularly suited for applications where light of a specific wavelength is used. The optical device is

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suited for use as a common optical disk substrate, of course, and also as a magneto-optical disk substrate because it has low birefringence for obliquely incident beams, as stated above, and has high heat resistance.

1 by Hitachi, Ltd.

Flexural properties: Measured in accordance with ASTM D-790.

Heat resistance: Expressed in terms of the glass
transition temperature measured by the
5 linear expansion coefficient method.

Intrinsic viscosity of polymer: Measured in chloroform at
25°C using Ubbelohde viscometers.

The kneading and pelletizing was carried out by
using a twin-screw extruder (Model TEX30-30BW-2V, supplied
10 by the Japan Steel Works, Ltd.).

The injection molding was carried out by using
an injection molding machine (Neomat Model 150/75 (75 ton),
supplied by Sumitomo Heavy Industries, Ltd.) and a mold for
producing a disk 120 mm across and 1.2 mm thick.

15 Example 1

Into a four-necked flask equipped with a thermometer, a reflux condenser, a dropping funnel, a stirrer, and a gas-introducing tube, were charged 90 g of xylene and 11 g of ethanolamine. To the mixture was added with stirring
20 88 g of a methanol solution containing 1.51 g of manganese (II) chloride, and oxygen was introduced therein at room temperature for 15 minutes. Subsequently, a solution containing 48.9 g of 2,6-xylenol and 49 g of xylene was charged in the flask, and while introducing oxygen the
25 reaction was allowed to proceed with stirring at 30°C, for 5 hours. The reaction mixture was poured into a large volume of methanol containing a small amount of hydrochloric

1 acid, and the precipitates formed were collected by
filtration, washed with methanol, then with water, and
dried to obtain 47.5 g of poly(2,6-dimethyl-1,4-phenylene)
ether having an intrinsic viscosity of 0.52 dl/g as
5 measured in chloroform at 25°C.

The polyphenylene ether resin and a polystyrene
resin (Tradename: Esbrite 8, general purpose grade, MFR 10,
supplied by Sumitomo Chemical Co., Ltd.) were blended
together in proportions shown in Table 1. This blend was
10 kneaded and pelletized through the extruder and then
injection-molded at a cylinder temperature of 320°C and a
mold temperature of 85°C to form disks 120 mm across and
1.2 mm thick.

Results of evaluating properties of these disks
15 are also shown in Table 1. The birefringence was measured
at a position 35 mm distant from the center of the disk.

Examples 2 to 4

Disks were prepared by following the procedure
of Example 1 but varying the blending proportions of the
20 poly(2,6-dimethyl-1,4-phenylene)ether and the polystyrene
resin. Results are shown in Table 1.

Table 1

	Example			
	1	2	3	4
Poly(2,6-dimethyl-1,4-phenylene)ether, (%)	30	35	40	50
Polystyrene, (%)	70	65	60	50
Light transmittance (%)	88	87	86	86
Birefringence (nm)	-50	-20	+20	+40
Water absorption (%)	0.1	0.1	0.1	0.1
Glass transition temperature (°C)	132	137	143	155

1 Comparative Example 1

Poly(2,6-dimethyl-1,4-phenylene)ether prepared in Example 1 was injection-molded alone.

The molding, infeasible under the same conditions as in Example 1, was carried out at a cylinder temperature of 350°C and a mold temperature of 130°C.

The birefringence value was as large as 100 nm or more in absolute value.

Comparative Example 2

10 The same polystyrene resin used in Example 1 was molded alone under the same conditions as in Example 1.

The birefringence was 100 nm or more in absolute

1 value and its distribution unevenness was greate.

Comparative Example 3

According to the ordinary method, a polycarbonate resin of average molecular weight about 15,000 was prepared
5 by the interfacial polycondensation of bisphenol A dissolved in methylene chloride with phosgene blown into the solution, wherein t-butylphenol was used for molecular weight control.

The obtained powdery resin was pelletized through
10 the extruder and injection-molded under the same conditions as in Example 1.

Disks thus obtained were found to cause large birefringence values of +100 nm and more.

Further, disks formed by molding the above pellets
15 at a cylinder temperature of 340°C gave a large birefringence value of 100 nm and showed a water absorption of 0.45. The glass transition temperature was 136°C.

The molding was further tried at a higher cylinder temperature, but at temperatures exceeding 340°C
20 the resin decomposed and hence was hardly molded.

Examples 5 to 16

Poly(2,6-dimethyl-1,4-phenylene)ethers of different intrinsic viscosity as shown in Table 2 were severally blended with a polystyrene resin (tradename: Esbrite
25 4-62A, supplied by Nihon Polystyrene Kogyo Co., Ltd.) in various proportions shown also in Table 2. These blends

1 were each kneaded, pelletized, and injection-molded under
the conditions of 320°C resin temperature, 100°C mold
temperature, and 1 second's injection period to prepare
optical disk substrates.

5 Table 2 shows values of birefringence of a
vertical incident beam and an oblique incident beam of 30°
caused by the thus obtained optical disk substrates. The
measurement position is expressed by the distance in the
radial direction from the center of the optical disk
10 substrate.

Table 2 also shows flexural properties and heat
resistance of plates prepared by press-forming of pelletized
resins above at 270°C.

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Table 2

Example No.	Composition			Birefringence (nm)						
	Poly(2,6-di-methyl-1,4-phenylene)ether	Vertical incident beam			30° oblique incident beam			At 25 mm	At 40 mm	At 55 mm
		Intrinsic viscosity	Parts	Parts	At 25 mm	At 40 mm	At 55 mm			
5	0.44	40	60	-15	-17	-18	-18	-18	-18	-19
6	0.44	44	56	+2	+1	+2	+3	+1	+1	+2
7	0.44	48	52	+19	+16	+15	+20	+18	+18	+17
8	0.40	46	54	-13	-10	-12	-9	-10	-10	-10
9	0.40	50	50	-1	-2	-2	0	-1	-1	-1
10	0.40	54	46	+10	+11	+12	+12	+12	+12	+12
11	0.35	55	45	-7	-8	-8	-8	-8	-8	-9
12	0.35	59	41	0	-1	-1	0	0	0	-1
13	0.35	63	37	+6	+6	+6	+8	+7	+7	+7
14	0.31	63	37	-4	-4	-4	-5	-5	-5	-5
15	0.31	67	33	+1	+1	+1	+1	0	0	0
16	0.31	70	30	+6	+5	+5	+6	+6	+6	+6

- cont'd -

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Table 2 (cont'd)

Flexural property				Heat resistance °C
Flexural strength Kg/cm ²	Flexural modulus Kg/cm ²	Maximum strain %		
1010	34,800	3.6		142
1180	35,200	4.2		148
1160	35,500	4.5		153
1010	35,300	3.2		149
1080	35,800	3.5		154
1100	35,800	3.7		159
700	34,200	3.0		150
720	34,500	3.1		155
750	34,600	3.3		159
430	35,000	1.6		149
410	34,900	1.4		154
420	34,800	1.5		158

CLAIMS:-

1. An optical device formed of a polymeric material comprising (i) polymer portions constituted mainly of aromatic vinyl monomer units and (ii) polyphenylene ether portions, characterized in that the average molecular weight of the polyphenylene ether portions is from 0.3 to 0.7, expressed in terms of the intrinsic viscosity as measured in chloroform at 25°C.
2. An optical device as claimed in claim 1 characterized in that the said average molecular weight of the polyphenylene ether portions is from 0.3 to 0.45.
3. An optical device as claimed in claim 1 or claim 2 characterised in that the polymer material comprises from 30 to 70% by weight of vinyl aromatic polymer and from 70 to 30% by weight of polyphenylene ether.
4. An optical device as claimed in claim 3 characterized in that the polymer material comprises from 40 to 55%, by weight of vinyl aromatic polymer and from 60 to 45% by weight of polyphenylene ether.
- 20 5. An optical device as claimed in any one of the preceding claims characterised in that the aromatic vinyl monomer is styrene and the polyphenylene ether is poly(2,6-dimethyl-1,4-phenylene)ether.

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6. An optical device as claimed in any one of the preceding claims characterised in that it is in the form of a magneto-optical disk substrate.



European Patent
Office

EUROPEAN SEARCH REPORT

0225801
Application Number

EP 86 30 9543

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
X, Y	US-A-4 373 065 (W.M. PREST Jr.) * Claims *	1-6	G 02 B 1/04 C 08 L 71/04 C 08 L 25/00
Y	FR-A-2 313 413 (GENERAL ELECTRIC) * Claims; page 15 *	1-5	

TECHNICAL FIELDS SEARCHED (Int. Cl.4)			
G 02 B C 08 L G 11 B			

The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
THE HAGUE	17-03-1987	DERAEDT G.	
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone	T : theory or principle underlying the invention		
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